8th Int. Symp. "Nanostructures: Physics and Technology"
St Petersburg, Russia, June 19–23, 2000

Carrier relaxation dynamics in self-assembled quantum dots

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Abstract. A novel technique to study carrier relaxation dynamics based on the artificial control of nonradiative losses by an external electric field is proposed. A clear evidence of phonon assisted relaxation as the main relaxation mechanism of hot electron-hole pairs in InP self-assembled quantum dots is found by the proposed method. Efficient one step relaxation processes with emission of acoustic and optical phonons are observed. These findings give new and important insight into the interaction of the electron-hole pairs in quantum dots with the phonon subsystem.

In this work we propose a new powerful method to study the carrier relaxation in self-assembled quantum dots (QDs) exploiting the phenomenon of photoluminescence (PL) suppression when an external electric field is applied to a sample containing the QDs. We found that under quasiresonant excitation, distinct resonances shifted from the excitation line by the LO or acoustic phonon energy appear in the spectra of the partially suppressed PL. We explain this phenomenon as a result of the competition between nonradiative losses and intraband carrier relaxation processes. The rate of nonradiative losses may be intentionally controlled by changing the applied bias. This opens up wide possibilities for the study of carrier relaxation dynamics.

We studied a few heterostructures with one layer of InP or InGaAs self-assembled QDs between $Ga_{0.5}In_{0.5}P$ or GaAs barrier layers, respectively. The samples was grown by gas source molecular beam epitaxy on an n^+ GaAs substrate. To study of the optical spectra of the sample in an electric field we provided the samples with a semi-transparent gold Shottky contact (thickness $\approx$20 nm) on the top surface and an ohmic contact on the back surface.

The PL bands of the QDs recorded under quasiresonant excitation at zero bias have a smooth profile without any sharp features as shown in Fig. 1(a). When a negative bias is applied to the sample surface, the intensity of the PL decreases and distinct sharp resonances appear in the PL spectra. Most prominent resonances are shifted from the excitation line approximately by the LO phonon energy of InP bulk crystal ($\hbar\omega = 43.5$ meV). PL excitation spectra reveal very similar behaviour with prominent 1LO and 2LO resonances at negative bias as is shown in Fig. 1(b). The PL spectra of the other studied samples reveal similar behavior (see Fig. 2).

All experimental data strongly suggest that the observed resonances are caused by fast relaxation of hot carriers with emission of LO or accoustic phonons rather than by phonon sidebands of the resonance PL or by resonant Raman scattering.

Our explanation of the observed phenomenon may be summarized as follows. The applied electric field activates a process of nonradiative losses of the QD excitation. Most likely cause of these losses is the tunneling of the hole from the QD into the barrier layer

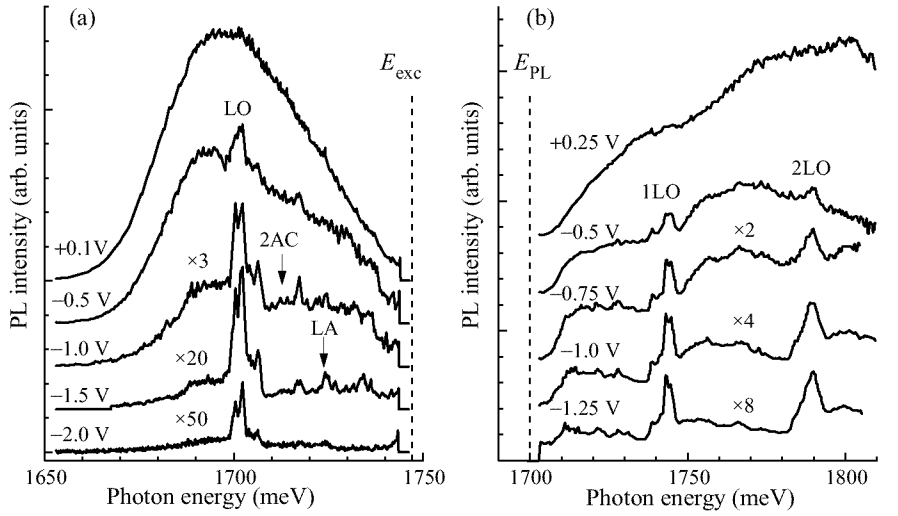

Fig. 1. Dependence of PL (a) and PL excitation (b) spectra on applied bias for the sample QDP1779 with InP QDs. Excitation power density 100 W/cm^2, $T = 2$ K. The excitation and detection photon energies are marked by dashed lines. Applied bias is shown near each spectrum.

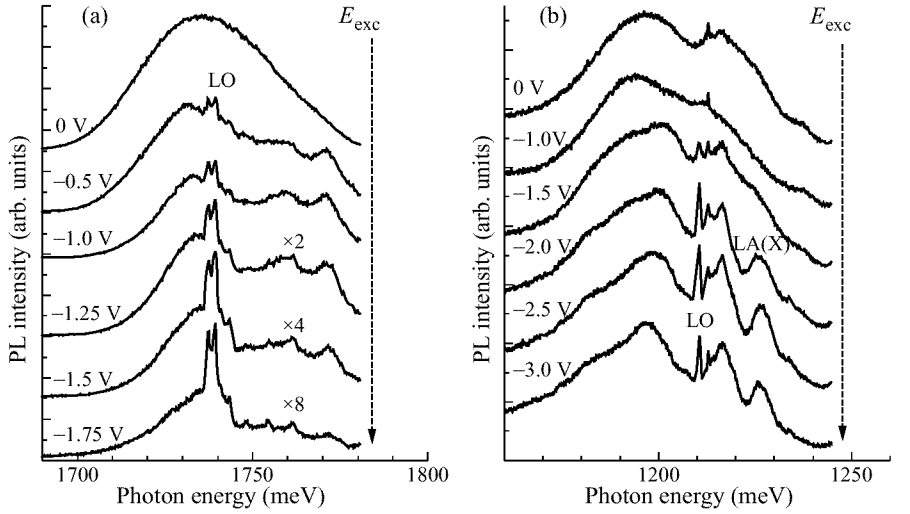

Fig. 2. PL spectra dependence on applied bias for sample QDP1778 with InP QDs (a) and 1142D with InGaAs QDs (b). Excitation photon energy marked by dashed arrows. Applied bias shown near each spectrum.

because of the depth of the potential well for holes is smaller than for electrons in such heterostructures. The details of this process is discussed elsewhere [ ]. The rate of the nonradiative process increases with the increase of negative bias. The tunneling from excited state competes with relaxation to the ground state. This leads to selective PL suppression because of different relaxation rates with emission of LO and acoustic phonons.

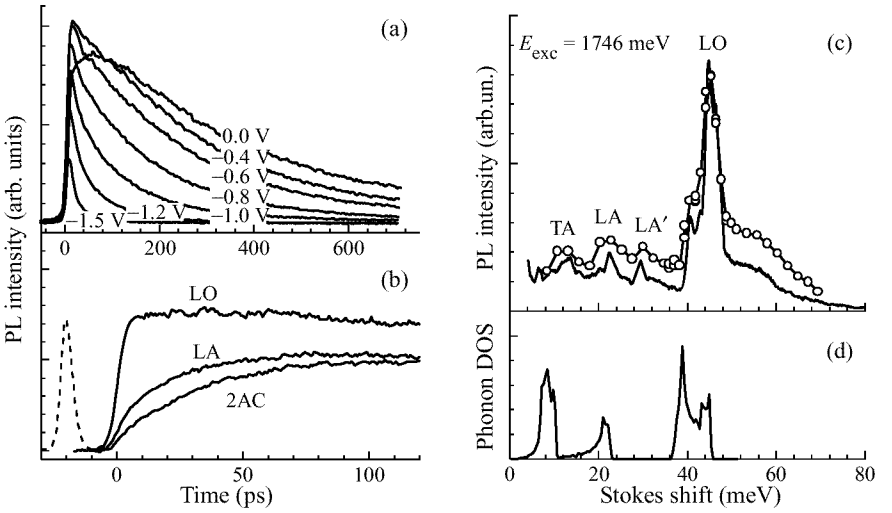

Fig. 3. (a) The PL kinetics at the LO resonance under various applied bias indicated on each curve. (b) Initial part of PL kinetics at zero bias at spectral points corresponding to one (LA) and two (2AC) acoustic phonon and LO phonon (LO) assisted relaxation marked in Fig. 1(a). The laser pulse (dashed curve) with full width at half maximum of 6 ps indicates the time resolution of the set-up. (c) The PL spectrum at $U_{\text{bias}} = -1.5$ V and cw excitation (solid line) and the time resolved PL spectrum reconstructed from kinetics data (circles) as described in the text. Solid line through the circles is a guide to the eye. "TA", "LA", "LA′", and "LO" mark the phonon resonances. (d) Phonon density of states of the InP crystal [2].

To verify the assumptions implicit in the above description, we performed the PL kinetics measurements for then samples with InP QDs. Kinetics for selected spectral points and their dependence on the applied bias for the LO resonance are shown in Fig. 3(a) and (b). As seen, the kinetics clearly demonstrate a shortening of the decay time with the increase of negative bias. So the nonradiative process forms some kind of time (or optical) gate for the PL. Initial (rise) part of the kinetics contains information about the relaxation rates. As seen from Fig. 3(b), it is very fast for LO resonance and limited by our time resolution of about 6 ps. In the spectral region formed by the acoustic phonon assisted relaxation (hereafter referred to as the acoustic region), the rise time is about 50 ps that is much slower than at the LO resonance.

It is clear from presented data that the manipulation of the nonradiative losses allows us to determine spectral dependence of the relaxation rate. To demonstrate it, we measured the PL kinetics at many spectral points at zero bias, then multiplied them by $e^{-\gamma_t t}$ to imitate the fast nonradiative losses and integrated. The time resolved PL spectrum reconstructed this way is shown in Fig. 3(c) together with the PL spectrum measured at $U_{\text{bias}} = -1.5$ V. As the estimated tunneling time at -1.5 V is shorter than the time resolution of our set-up, $\tau_s = 6$ ps, we have used $\gamma_t = 1/\tau_s$. The excellent agreement between the two spectra clearly demonstrates the consistency of our description of the PL process. At the same time, the spectral and effective time resolutions in the PL spectrum of the biased sample are better than that achievable in kinetics experiments.

A few important statements about relaxation mechanisms and relaxation rates can be derived from the experimental data and the analysis presented above.

The main relaxation mechanism of hot carriers in QD's when only one electron-hole pair is created in the QD is the phonon assisted relaxation. Different relaxation channels are possible. Electron and hole may relax separately or together, mediated by the Coulomb interaction, by emission of phonons. The PL kinetics for the LO resonance shows mainly a fast rise component. From this kinetics it follows that single LO phonon emission contributes more than 70% of the PL at the LO resonance energy.

The relaxation time with emission of LO phonons lies within the range 0.6 ps $< \tau_{LO} <$ 6 ps. The lower bound is derived from fitting the peaks in the LO resonance by Lorenzians. The upper bound is defined by the time resolution of the direct kinetics measurements.

Acoustic phonon resonances are observed in the spectra of the partially suppressed PL. A comparison of the PL spectrum with the phonon density of states of InP crystal (see Fig. 3(d)) allows us to attribute the resonances to the transverse acoustic (TA) and longitudinal acoustic (LA) peaks in the phonon density of states (DOS) of InP and $Ga_{0.5}In_{0.5}P$. The rising kinetics of the PL in these peaks are faster than in the rest of the acoustic part of the spectrum. This observation is evidence of efficient relaxation of electron-hole pairs with emission of high frequency acoustic phonons, in stark contrast with theoretical predictions [3].

In conclusion, we developed a novel method for the study of carrier relaxation in QD's based on the artificial control of the nonradiative losses by an external electric field. The nonradiative process with a controllable rate gives rise to an effective optical gate with variable duration for the PL of the QD's. This method allows one to study the spectral dependence of the carrier relaxation rates with high spectral and time resolution at a low power density of optical excitation. Our results clearly demonstrate that the main relaxation mechanism in InP self assembled QD's is the phonon assisted relaxation. Clear evidence of efficient one step relaxation with emission of high frequency acoustic phonons is found. This observation poses a new principal problem for theoretical analysis.

References

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